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Colossal spin Hall effect in ultrathin metallic films

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We predict spin Hall angles up to 80% for ultrathin noble-metal films with substitutional Bi impurities. The colossal spin Hall effect is caused by enhancement of the spin Hall conductivity in reduced sample dimension and a strong reduction of the charge conductivity by resonant impurity scattering. These findings can be exploited to create materials with high efficiency of charge to spin current conversion by strain engineering.

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An efficient transformation of charge into spin current is the key issue for future applications of the spin Hall effect (SHE) [1,2] in spintronic devices. The figure of merit for this phenomenon is described by the spin Hall angle (SHA). For a long time, its largest value was 11%, measured in Au [3]. Subsequently, several studies revealed systems that provide SHAs of comparable magnitude, the so-called *giant* SHE [3]. Among them, a SHA of -12% to -15% was measured in highly resistive β -Ta [4]. Even larger values of 30% to 33% were obtained by an experimental study of β -W thin films [5]. Both results have been qualitatively predicted evaluating a tight-binding model [6]. In addition, a SHA of -24% has been reported for thin-film Cu(Bi) alloys [7]. The large magnitude of the SHA for a copper crystal with Bi impurities was predicted by first-principles calculations relying on the skew-scattering mechanism [8]. The existing results inspire both theorists and experimentalists to search for new routes to synthesize materials with even larger SHAs.

In this Rapid Communication we show that the SHA caused by impurities in ultrathin metallic films can reach values up to 80%. We concentrate on the skew scattering as the responsible mechanism for the SHE in dilute alloys [9,10]. The parameter we are going to optimize is the spin Hall angle,

$$\alpha = \frac{\sigma_{yx}^s}{\sigma_{xx}}, \quad (1)$$

defined as the ratio of the spin Hall conductivity (SHC) σ_{yx}^s to the longitudinal charge conductivity σ_{xx} . This ratio is dimensionless since both conductivities are expressed in the same units [8,11,12]. For the skew-scattering mechanism, both depend inversely on the impurity concentration, which leads to an α that is concentration independent. Consequently, this quantity is ideally suited for comparison with experiment.

In our previous study [11] we showed that the SHE induced by Pt impurities in Au(111) films can be significantly enhanced for a reduced film thickness. In particular, for the one-monolayer (ML) films, a large SHA of -13% to -18% was obtained for the case of Pt adatoms. Generally, the reduced coordination number and the associated strong potential gradients enhance the effective spin-orbit coupling (SOC). In addition, a single-sheeted Fermi surface of 1-ML noble-metal films leads to the absence of interband

scattering causing the giant SHE via increased anisotropic scattering [11]. However, such a strong effect was caused by adatoms, which leaves the feasibility of corresponding systems for stable practical devices questionable. On the other hand, substitutional Bi impurities in bulk Cu cause a giant SHE as well. In this case the large valence difference between impurity and host atoms substantially increases potential scattering as well as effective SOC. Moreover, Bi impurities are strong p scatterers [7,12,13], which typically enhance the SHC [8]. The idea of this Rapid Communication is to combine both facts and screen the influence of Bi impurities in ultrathin noble-metal films. Accordingly, we focus on the 1-ML films, considering Bi as an adatom or substitutional impurity. Figure 1 shows the general setup for our calculations.

The first-principles electronic structure of the host as well as the impurity system is obtained by means of the relativistic Korringa-Kohn-Rostoker Green's function method [14]. The transport properties are calculated within the semiclassical theory via a solution of the Boltzmann equation, as it was developed originally for bulk crystals [8,15] and later extended to the case of two-dimensional (2D) systems [11].

The corresponding results obtained by this approach are presented in Fig. 2, where the SHA, the SHC, and the charge conductivity are shown for 1-ML (111) and (001) noble-metal films. Throughout this Rapid Communication, the impurity density chosen for the films corresponds to the 1 at. % impurity concentration used for the bulk crystals. First, nearly all values of the SHA shown in Figs. 2(a) and 2(b) are drastically increased in comparison to the related bulk systems with $\alpha < 10\%$ [16]. The SHAs for the (111) films are even larger than the values reported in Ref. [11] for the corresponding hosts with Pt impurities ($\alpha < 19\%$). Thus, combining the reduced coordination number of the impurity with the large valence difference between impurity and host atoms seems to be the proper way to enlarge the SHA [17]. In detail, the enhancement of the SHA in comparison to the bulk results is

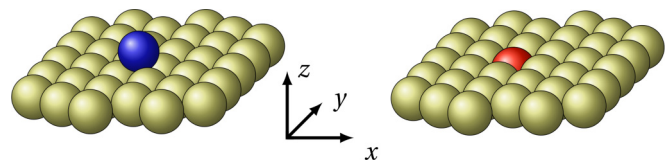


FIG. 1. (Color online) Geometry of the considered systems shown for a freestanding 1-ML fcc (111) film with (left) an adatom or (right) a substitutional impurity.

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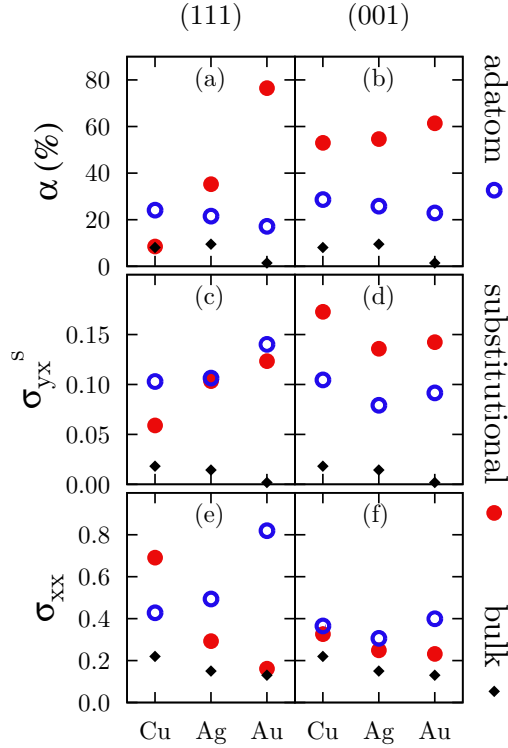


FIG. 2. (Color online) The spin Hall angle α , the spin Hall conductivity σ_{yx}^s , and the charge conductivity σ_{xx} for 1-ML (111) and (001) Cu, Ag, and Au films with Bi as adatoms (blue open circles) and substitutional impurities (red solid circles). For comparison, the corresponding bulk values [16] are shown as black diamonds. The conductivities are given in units of $(\mu\Omega \text{ cm})^{-1}$, while α is shown as a percentage.

determined solely by the SHC, which is strongly increased in all systems, as shown in Figs. 2(c) and 2(d). By contrast, the charge conductivity alone would cause a reduction of α since it is also increased in comparison to the bulk values but enters Eq. (1) in the denominator.

Another striking feature is the strong dependence of the SHA on the host material for substitutional Bi impurities in (111) films, culminating in the 80% value for Au. In contrast to the other configurations, σ_{yx}^s and σ_{xx} change for this case in the opposite direction, going from Cu via Ag to Au [see Figs. 2(c) and 2(e)]. As a result, both conductivities interact to amplify the corresponding SHA shown in Fig. 2(a). However, the influence of the charge conductivity is stronger. Within the (111) films σ_{xx} is reduced by a factor of 4, changing the host from Cu to Au. The particularly small charge conductivity of Au(111) films with substitutional Bi impurities finally causes the colossal effect. Therefore, the following detailed analysis will focus on substitutional impurities.

In order to get a deeper insight into the underlying mechanism of the colossal SHE, we investigate the energy dependence of the obtained quantities. The corresponding values for the SHA and its constituents, σ_{yx}^s and σ_{xx} , are shown in Fig. 3. In all cases α is strongly energy dependent. Here, it is important to mention that the considered energy interval is so small that the topology of the single-sheeted 2D Fermi surface of the 1 ML films [11] remains unchanged, except for

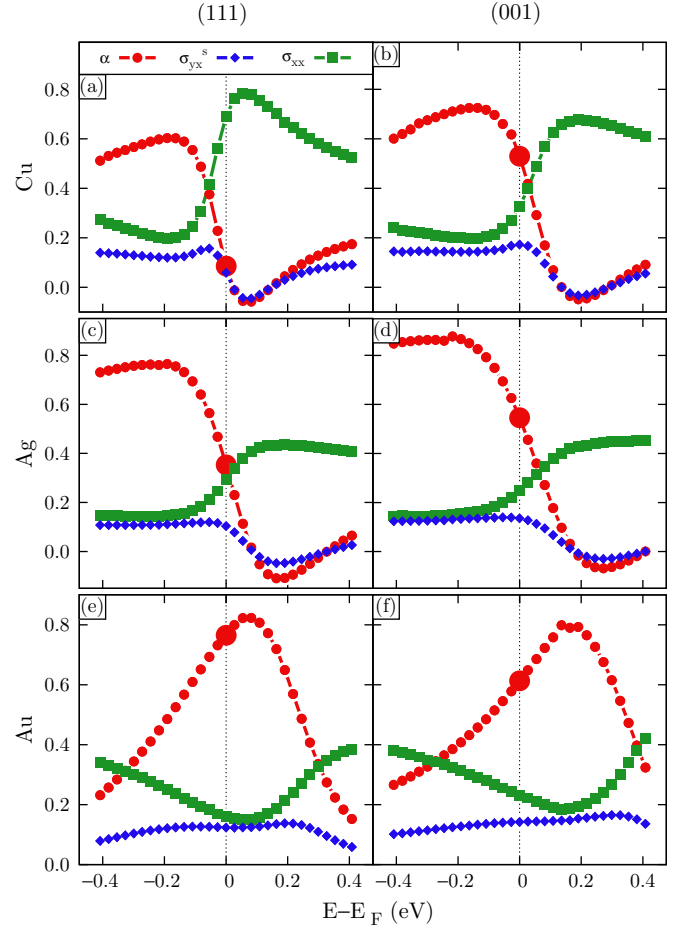


FIG. 3. (Color online) The results of transport calculations performed at different energies around the Fermi level (E_F) are shown for 1 ML (111) and (001) Cu, Ag, and Au films with substitutional Bi impurities. The corresponding α (red filled disks), σ_{yx}^s (blue diamonds), and σ_{xx} (green squares) are displayed and $\alpha(E_F)$ is highlighted with a larger disk. All quantities have absolute values in the same order of magnitude but different units: the conductivities are in $(\mu\Omega \text{ cm})^{-1}$ while α is dimensionless and has to be multiplied by 100 to get the values in percent.

the Au (001) film where small parts of the second band arise below 0.14 eV.

Analyzing the contributions to the SHA from the charge and spin Hall conductivity separately, one can see differences depending on the host material. While for Au the energy dependence of α is determined almost solely by the charge conductivity, σ_{yx}^s and σ_{xx} play a comparable role for Cu and Ag. Nevertheless, a certain correlation between the two conductivities exists for all considered systems. Namely, the SHC increases with decreasing charge conductivity and vice versa, which is most obvious for the Ag films. This is related to the fact that, generally, stronger scattering should reduce σ_{xx} but enhance σ_{yx}^s [11]. However, the situation in real systems can differ from such a simplified picture. For instance, the pronounced maximum of the SHA in the Au films is determined merely by the corresponding minimum of the charge conductivity. For both (111) and (001) Au films small changes in energy strongly affect σ_{xx} , while σ_{yx}^s remains

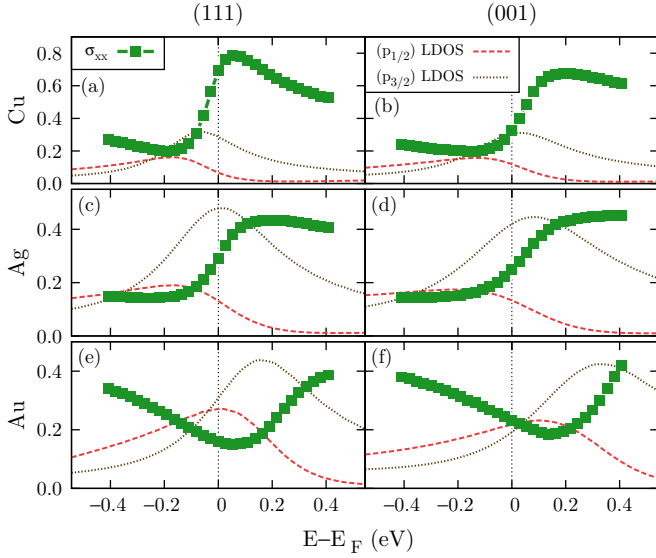


FIG. 4. (Color online) The contributions of the relativistic $p_{1/2}$ channel (dashed red line) and $p_{3/2}$ channel (dotted brown line) to the impurity local density of states (LDOS) are shown in comparison to the energy-dependent charge conductivity σ_{xx} (green squares) for 1-ML (111) and (001) Cu, Ag, and Au films with substitutional Bi impurities. The charge conductivity and the impurity LDOS are plotted in units of $(\mu\Omega \text{ cm})^{-1}$ and states/eV, respectively.

almost constant. In the case of the 1-ML Au(111) film the minimum of the charge conductivity occurs in the vicinity of the Fermi level E_F , which causes the colossal SHA shown in Fig. 2(a). Thus, going from bulk Au to the 1-ML Au(111) film, the giant SHE occurs since the SHC is increased due to the enhancement of the SOC and the absence of interband scattering [11]. Finally, the SHA is further enhanced through the suppression of the charge conductivity. These are the ingredients of the reported colossal SHA.

An enhancement of the SHA due to resonant scattering follows from the Fert-Levy model for the SHE [18], where the SHC is proportional to the ratio of the spin-orbit constant and the resonance width. In addition, the resonant scattering reduces the charge conductivity, as demonstrated in Fig. 4, which presents the impurity local density of states (LDOS). As mentioned above, Bi impurities are known to be strong p scatterers. Because of SOC the p level is split into $p_{1/2}$ and $p_{3/2}$. The minimum of the charge conductivity is obviously correlated to the maximum of the $p_{1/2}$ LDOS of the Bi impurity (Fig. 4).

Although the correlation between the $p_{1/2}$ impurity LDOS and the charge conductivity is obvious, the underlying mechanism is quite complex. For the considered films the energy dependence of σ_{xx} , with its minimum related to the maximum in the $p_{1/2}$ impurity LDOS, is predominantly provided by the vertex corrections [19]. In the Boltzmann equation they are represented by the so-called *scattering-in* term [20], which is the source of the considered skew-scattering contribution to the SHC [8]. The Boltzmann equation, which is an integral equation, is solved iteratively in our approach [19]. Therefore, it is difficult to provide a simple connection between the impurity LDOS and the charge conductivity.

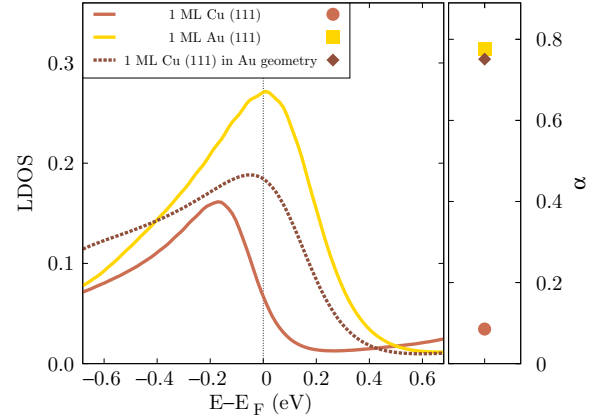


FIG. 5. (Color online) (left) The $p_{1/2}$ local density of states at a substitutional Bi impurity atom in a 1-ML fcc (111) film of Cu [solid copper (dark gray) line], Au [solid yellow (light gray) line], and Cu with the gold lattice constant (dashed brown line). (right) The corresponding SHA calculated at E_F for Bi in Cu (circle), Au (square), and Cu with the gold lattice constant (diamond).

Nevertheless, the strong influence of the vertex corrections is clearly related to the resonance scattering. Indeed, the well-pronounced peaks of the impurity LDOS near E_F , shown in Fig. 4, are missing in the case of substitutional Bi atoms in bulk Au [19] and Cu [21]. As a consequence, the energy dependence of the charge conductivity for bulk crystals is practically unaffected by the scattering-in term [19].

With this microscopic picture in mind, it is possible to understand the host dependence of α for the two surface orientations. As illustrated by Fig. 3, the general energy dependence $\alpha(E)$ is extremely similar between (001) and (111) films for each host material. However, in the (001) case the reduced coordination number causes a smaller charge density. As a result, the related impurity resonance is shifted to higher energies with respect to the Fermi level. The conductivity minimum is shifted accordingly. In addition, strong changes in σ_{xx} at energies around E_F cause strong variations of α . Thus, it is somewhat accidental that the SHA shown in Fig. 2 is nearly the same for the (001) films, while it varies strongly for the (111) films. Shifting the Fermi level of the (001) films slightly to higher energies would simulate the situation of the (111) surface orientation.

This finding can be employed to optimize the SHA. The only condition required is to fix the $p_{1/2}$ impurity resonance at the Fermi level. A possible opportunity is strain engineering of the film grown on an appropriate substrate. To prove this assumption, let us choose the Cu(111) film with the smallest SHA among all the considered systems, as indicated by Figs. 2(a) and 2(b). According to Fig. 4, the peak of the corresponding $p_{1/2}$ impurity LDOS is below E_F . Following the discussion above, we can assume that an increase in the host lattice constant should provide a shift of the impurity resonance towards the Fermi level. Figure 5 shows that the desired condition can be fulfilled if the lattice constant of Au is used for the hypothetically strained Cu(111) film. The increase of the lattice constant by about 13% from $a_{\text{Cu}} = 3.615 \text{ \AA}$ to $a_{\text{Au}} = 4.078 \text{ \AA}$ shifts the maximum of the $p_{1/2}$ impurity LDOS close to E_F . This, in return, leads to the colossal SHA, as

shown in the right panel of Fig. 5. Of course, the considered change in the lattice constant is quite strong. Nevertheless, the presented results indicate a new route to design materials with large α .

A substrate is needed in general since the freestanding 1-ML films considered in our study are quite artificial from a practical point of view. To have more realistic systems for the observation of the colossal SHE, these films need to be grown on an insulating substrate that meets the required conditions discussed above. Those requirements can be enforced by the aforementioned strain (or constriction) as well as induced electrostatic forces affecting the relative position of the Fermi level with respect to the impurity states. However, the considered skew-scattering mechanism is strongest for host electron spins pointing out of the plane, while the Rashba-type SOC induced by a substrate would force them to be oriented within the film plane [22]. In order to ensure the desired spin orientation, a symmetric quantum well structure with insulator/metal film/insulator could be utilized.

In addition, structural relaxations around impurities should occur in the considered systems due to the large differences between Bi and host atom sizes. The influence of this effect was considered in Ref. [12] for the Cu(Bi) alloy. It was shown that the relaxation of 5.6% obtained for the nearest neighbors in a Cu host around Bi impurities leads to a relative reduction

of the SHA of about 20%. Taking into account the much larger size of Au atoms as well as the related lattice constant, which is 13% larger than in Cu, we expect a much weaker structural relaxation in the case of Bi impurities in gold. From this perspective, we do not anticipate that structural relaxation will significantly affect the obtained colossal SHE.

In summary, we predict a colossal spin Hall effect related to spin Hall angles up to 80%, which are obtained for ultrathin noble-metal films with substitutional Bi impurities. This strong effect occurs for systems where the giant SHE, caused by the reduced dimension of the samples, is further amplified by a minimal charge conductivity caused by resonant scattering. This condition is achieved if the impurity $p_{1/2}$ resonance state is pinned at the Fermi level. The required resonant scattering can be tuned by strain engineering, which deposits films on an appropriate substrate. Our findings offer a new route to design materials with a very efficient conversion of charge current into spin current.

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